Strain Relaxation and Strengthening Mechanisms

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X-Ray Diffraction Analysis and Modeling of Strain Induced Thermal Cycling in a Thin Aluminum (011) Bicrystal Film

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ABSTRACT

Heteroepitaxial films of aluminum bicrystals grown on silicon provide a model system in which to study plasticity in polycrystalline metal thin films. For the bicrystal films, dislocations are confined to move on two different slip plane orientations because of the orientation of the crystals on the substrate. *In-situ* transmission electron microscopy (TEM) observations during thermal cycling have shown two threshold temperatures for dislocation motion on cooling. A simple model uses the resolved shear stress on the possible slip planes to explain the TEM observations. Mechanisms responsible for the dislocation behavior are studied *in-situ* during thermal cycling between room temperature and 450ºC with x-ray diffraction. The strains are determined using a $\sin^2(\psi)$ analysis at each temperature. Direct comparisons are made between the TEM observations, the model and x-ray diffraction results.

INTRODUCTION

Plasticity in thin metal films is a subject of intense research. There are fundamental questions about size effects on dislocation motion and the importance of dislocation-dislocation interactions on the strength of thin films. Heteroepitaxial films of aluminum bicrystals grown on silicon provide a model system in which to study thin film plasticity. We report here on the plasticity of Al (011) bicrystal films grown on Si (001).

Dislocation structures have been observed *in-situ* in a 100 nm film using transmission electron microscopy (TEM) during thermal cycling by Stach *et al.* [1] (Figure 1). When cooling from a high temperature, there are two apparent threshold temperatures for dislocation movement. The first occurs around 275ºC and leaves misfit dislocations only along the

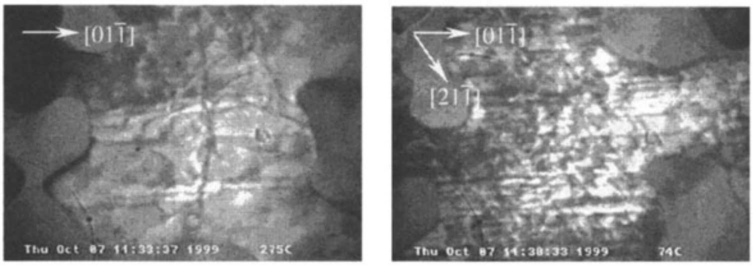

Figure 1. *TEM images taken* in-situ *while cooling from 450ºC. Two threshold temperatures are observed. At 275ºC (left) misfit dislocations are generated along the* [01$\bar{1}$] *direction and at 74ºC misfit dislocations are generated along both the* [01$\bar{1}$] *and the* [21$\bar{1}$] *directions.*

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$[01\bar{1}]$ direction. The second occurs around 74ºC and corresponds with a "burst" of dislocation movement that leaves misfit dislocations along the $[01\bar{1}]$ and $[21\bar{1}]$ directions.

In order to better understand this behavior, we have performed *in-situ* x-ray diffraction (XRD) measurements during thermal cycling. A simple model based on the existing slip systems and a channeling criterion for dislocation movement is presented and compared with the experimental observations.

EXPERIMENTAL PROCEDURE

A 100 nm heteroepitaxial aluminum bicrystal film was deposited by evaporation on a [001] silicon substrate at 280ºC. Previous studies have shown that under such experimental conditions Al grows heteroepitaxially with only two in-plane orientations: $(011)_{Al}$ $[100]_{Al}$ // $(001)_{Si}$ $[110]_{Si}$ or $(011)_{Al}$ $[01\bar{1}]_{Al}$ // $(001)_{Si}$ $[110]_{Si}$. The deposition method is described elsewhere [2]. TEM cross sectional views of similar samples are shown in Figure 2.

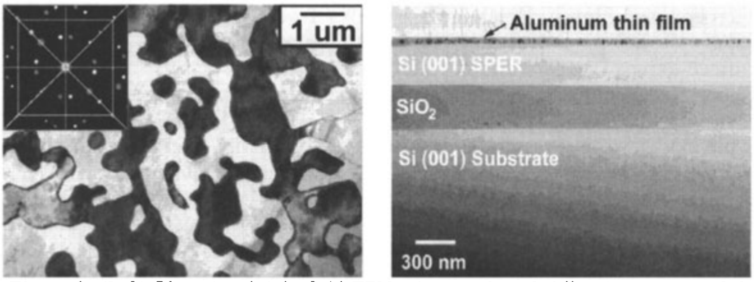

Figure 2. *(Left) Plan view bright field TEM micrograph and diffraction pattern of the aluminum film microstructure (silicon substrate removed). (Right) Bright field cross sectional TEM micrograph of a poly-crystalline aluminum layer as deposited.*

XRD measurements were performed *in-situ* during thermal cycling at the Cornell High Energy Synchrotron Source (CHESS) on the C1 beam line with an energy of 8 keV (0.155 nm). An Ordela 1100X position-sensitive proportional counter was used to record the 2θ peak position for three planes in the $[01\bar{1}]$ zone axis, (133), (222) and (422). The sample was placed on a water-cooled heating stage enclosed by a beryllium tube and ultra high purity nitrogen was flowed through the chamber during testing at 90 mL/min. The film was thermally cycled to 450ºC with an average heating/cooling rate of approximately 5º/min. The temperature was held constant for approximately 5 minutes at each of 29 measurement temperatures.

RESULTS AND DISCUSSION

The presence of two threshold temperatures observed in TEM during the cooling cycle is easily accounted for based on the resolved shear stress on the slip planes present in the aluminum (011) bicrystal. The mechanisms responsible for the dislocation behavior are studied using a quantitative measurement of the strain in the film as a function of temperature by XRD.

Anisotropic plasticity model

For an aluminum (011) Face-Centered Cubic (FCC) crystal there are four {111} slip planes oriented with respect to the film plane as shown in Figure 3. Two adopt inclined orientations 35º from the film surface and lie in the $[01\bar{1}]$ zone axis. The other two are perpendicular to the film surface and intersect it along a $\langle 21\bar{1}\rangle$ type direction.

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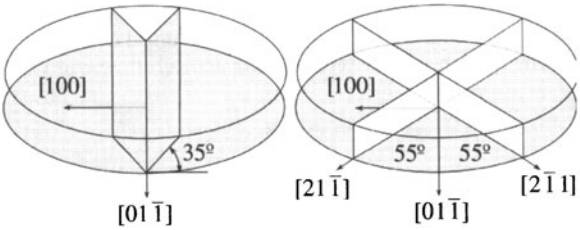

Figure 3. *Orientation of the inclined (left) and perpendicular (right) {111} slip planes in a single grain of the Al (011) film.*

A quantitative analysis of the stress required to move a dislocation on its slip plane is based on the channeling stress argument advanced by Freund and Nix [3, 4]. Following Freund's formulation, the channeling criterion is given by

$$\sigma^*_{RSS} = \frac{\sin(\beta)}{bh}\left[\frac{\mu_r b_e^2}{4\pi(1-\nu)}\left[\ln\left(\frac{2h}{r_0}\right)+\frac{1}{2}\cos 2\beta-\frac{1-2\nu}{4(1-\nu)}\right]+\frac{\mu_r b_s^2}{4\pi}\ln\left(\frac{2h}{r_0}\right)\right] \tag{1}$$

where

$$\mu_r = \frac{2\mu_f\mu_s}{\mu_f+\mu_s}, \tag{2}$$

the edge and screw components of the Burgers vector are $b_e = \pm b\,cos(\alpha)$ and $b_s = \pm b\,sin(\alpha)$ respectively, and the angles $\alpha = 60^{\circ}$ and 30° apply to the inclined and perpendicular planes, respectively. σ_{RSS}* is the critical resolved shear stress for dislocation motion including channeling effects. The film thickness is defined by h, β is the angle between the slip plane normal and the film normal, the shear modulus in the film and substrate are $\mu_f = 28$ GPa and $\mu_s =$ 76.9 GPa, respectively, the Burgers vector is $b = 0.286$ nm, Possion's ratio is $\nu = 0.37$, and the inner cutoff radius is $r_0 = b/2$.

For an equal-biaxial stress state, the resolved shear stress on the inclined planes is given by

$$\sigma_{RSS} = \frac{\sigma_{[100]}}{\sqrt{6}} \tag{3a}$$

and on the perpendicular planes is given by

$$\sigma_{RSS} = \frac{\sigma_{[01\bar{1}]}-\sigma_{[100]}}{\sqrt{6}}. \tag{3b}$$

Because the resolved shear stress for the perpendicular planes depends on the *difference* between the stresses in the [100] and the $[01\bar{1}]$ directions, there can only be a resolved shear stress on the perpendicular planes for an anisotropic stress state. The channeling criteria for slip on both the inclined and perpendicular planes is calculated from Equation (1) and gives

$$\sigma^*_{[100]} = 140MPa \text{ and } \sigma^*_{[01\bar{1}]}-\sigma^*_{[100]} = 186MPa \tag{4}$$

respectively.

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The strain and stress behavior as a function of temperature can now be modeled quantitatively, as shown in Figure 4. The strain is assumed to be zero at 450ºC. As the film cools, differential thermal expansion between the film and substrate will yield a total strain given by

$$\varepsilon = \int_{T_0}^{T} [\alpha_s(T) - \alpha_f(T)]dT \; . \tag{5}$$

Here α_s and α_f are the thermal expansion coefficients of the substrate and film, respectively. Initially, the strain will be completely elastic and produce a stress in the film given by the elastic constants of aluminum. The small differences in slope for the initial stress-temperature data result from the mild elastic anisotropy of aluminum. When the stress in the [100] direction reaches the channeling criterion for the inclined slip planes, dislocations will move on these planes. If we assume that there are no dislocation-dislocation interactions, the stress in this direction will not increase further. As the film continues to cool, the stress in the $[01\bar{1}]$ direction, however, does continue to increase since dislocation motion on the inclined planes will not relax the stress in the $[01\bar{1}]$ direction. This creates an anisotropic stress state, which, in turn, affects the strain in the [001] direction according to Poisson's effect. The strain in the $[01\bar{1}]$ direction continues to increase elastically. Finally, when the difference in stress for the two directions is large enough, the channeling criterion for the perpendicular planes is reached and dislocations can move on these planes.

The model predicts two threshold temperatures where dislocations should move at 385ºC and 270ºC on the inclined and perpendicular planes, respectively. These temperatures clearly do not agree with the observations made by TEM. The temperatures at which dislocation motion is observed in TEM are much lower (Fig 1). This implies either that much higher stresses than the channeling stresses are needed for dislocation motion to occur, or that plastic deformation is taking place before the channeling criteria are reached. In either case, dislocation motion would not be observed until lower temperatures.

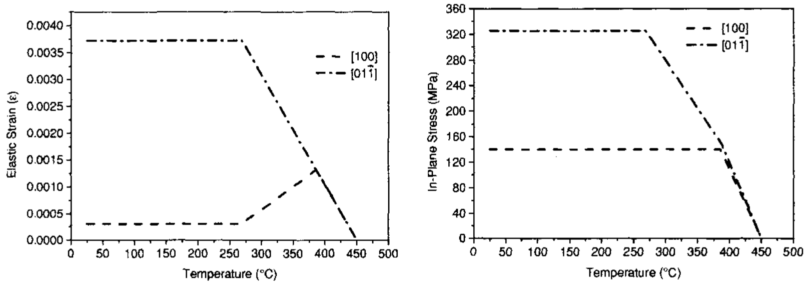

Figure 4. *Anisotropic plasticity model of the strain and stress as a function of temperature based on the resolved shear stresses and a channeling criteria for the slip planes.*

X-ray diffraction

X-ray measurements were made during each of two thermal cycles to 450ºC. Diffraction peaks from each of the three measured planes were fit with a gaussian function to determine the 2θ peak position, which was then converted to a lattice parameter using Bragg's Law and the crystal geometry. The measurements include only one of the grain orientations and represent an average over all of the grains illuminated by the x-ray beam. A $\sin^2(\psi)$ analysis [5] was used to find the strain at each temperature. The unstrained lattice parameter used for the analysis was that found at $\psi_0 = 45^\circ$ in the analysis. The in-plane strain in the [100] direction, $\varepsilon_{[100]}$, as a function of temperature for the second cycle is plotted in Figure 5.

The expected thermoelastic behavior calculated from Eq. 5 using temperature dependant thermal expansion coefficients [6, 7] is also plotted in Fig 5. The strain in the film follows this thermoelastic behavior, shown as a dotted line, on heating up to 200ºC but deviates from the thermoelastic line while the film is still in tension. Similar results have been observed in thin copper films and attributed to an energy storage mechanism [8, 9]. The film continues to deform plastically up to the maximum temperature of 450ºC.

On cooling, plasticity occurs almost immediately from 450ºC; certainly before the channeling criterion on the inclined planes has been reached. Observations made in TEM suggest that there is insufficient dislocation motion before 275ºC to account for the plasticity measured by XRD. This stress relaxation may possibly be explained by diffusional relaxation or by an energy storage mechanism similar to that seen on heating.

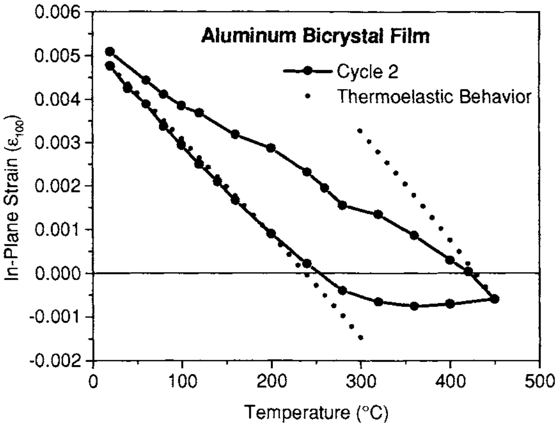

Figure 5. *XRD Results for $\varepsilon_{[100]}$ as a function of temperature for the second thermal cycle.*

At around 320ºC the slope of the strain-temperature data decreases temporarily. This temperature coincides well with TEM observations of the threshold temperature for movement of dislocations on inclined slip planes, as well as the model prediction of the channeling strain, $\varepsilon_{[100]} = 1.32\times10^{-3}$. The agreement between the TEM observations, model and XRD measurements suggests that there is large scale dislocation motion at the first threshold temperature.

Below 280ºC, the slope of the strain-temperature data increases sharply, nearly to that of the thermoelastic line and then decreases gradually until about 100ºC. In this region only a limited amount of dislocation activity is observed in TEM. Nonetheless the strain-temperature data indicate that plastic deformation and strain hardening are both occurring. We suggest that the strain hardening may be due to dislocations on the two inclined planes (only one of which is observed in TEM) interacting. No further features that can be correlated with dislocation activity are observed in the strain-temperature data. Below about 100ºC, the slope slowly rises until room temperature is reached. The fact that dislocation activity on the perpendicular planes is not observed in TEM until 74ºC is consistent with these observations. Since the channeling criterion for the perpendicular planes depends on the *difference* between stresses in the $[01\bar{1}]$ and [100] directions and since the stress in the [100] direction increases due to strain hardening, much higher total strains must be applied to activate the perpendicular planes.

It is worth noting that the number of dislocations that are generated in the observed area in Fig. 2b during the low temperature burst are enough to cause significant strain relaxation. The fact that no corresponding relaxation is observed in the XRD data (Fig. 5) suggests that such bursts must be local phenomena occurring at different temperatures in different regions, or perhaps only occurring in a few locations in the film.

CONCLUSIONS

Al (011) bicrystal films have been studied *in-situ* during thermal cycling by TEM and XRD. The dislocation activity observed in the TEM experiments can be explained based on a model of the resolved shear stress on the available slip planes. XRD experiments show that plasticity occurs before the channeling criterion for the inclined planes is reached. Strain hardening occurs after the first threshold temperature so that the stress anisotropy and thus the expected threshold temperature for initiation of dislocation motion on perpendicular slip planes are lower than predicted. Despite the high elastic isotropy in these films, plastic deformation appears to be very inhomogeneous. Further studies of this and other model systems will provide further insight regarding stress levels and plasticity in thin metal films.

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Direction Dependent Grain-Interaction Models for the Diffraction Stress Analysis of Thin Films

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ABSTRACT

The well-known grain-interaction models for the description of the macroscopic elastic behaviour of polycrystalline specimens, for example the models due to Voigt and Reuss, may be applied to bulk specimens, but they are generally not suitable for thin films because they imply macroscopic elastic isotropy of the body. A thin film is usually at most transversely elastically isotropic, even in the absence of texture. A recently elaborated, alternative model for grain-interaction in thin films, adopting grain-interaction assumptions first given by Vook and Witt (*J. Appl. Phys.* 7, 2169 (1965)), is able to predict the transversely isotropic elastic behaviour. Although this model is more appropriate for thin films than traditional models, it still imposes extreme grain-interaction assumptions, which are in general not compatible with the true elastic behaviour of real specimens. In this paper a more general approach to grain-interaction in thin films is proposed.

INTRODUCTION

So-called diffraction elastic constants are needed for the evaluation of diffraction stress measurements [1,2]. Generally, diffraction and macroscopic, mechanical elastic constants of polycrystals are calculated from single crystal elastic constants by adopting a grain-interaction model, describing the distribution of stresses and strains over the crystallographically differently oriented crystallites in a polycrystalline aggregate. The most widely used models are the Voigt [3], Reuss [4], Neerfeld-Hill [5,6] and Eshelby-Kröner [7,8] models. Devised for bulk specimens, these models imply macroscopically isotropic elastic behaviour for non-textured polycrystals. However, thin films are usually not macroscopically isotropic but exhibit only transverse isotropy along the plane of the film. Only recently van Leeuwen *et al.* [9] demonstrated that a grain-interaction model, adopting grain-interaction assumptions first formulated by Vook and Witt [10], can explain experimental findings (curved $\sin^2\psi$-plots; see below) for an untextured Ni film that are incompatible with the traditional grain-interaction models (see also [11]). Although the grain-interaction assumptions of Vook and Witt are more appropriate for thin films, still extreme (unphysical) constraints are imposed (see below). A more general approach to grain-interaction in thin films is proposed in this paper and applied to the diffraction analysis of stress in a fibre-textured copper layer.

BASIS OF DIFFRACTION STRESS ANALYSIS

A plane, rotationally symmetric state of residual stress is often met in thin films. Then, only one independent stress tensor component, the in-plane stress $\sigma_{//}$, has to be determined. For the diffraction stress analysis of a macroscopically elastically anisotropic specimen (e.g. a specimen

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exhibiting crystallographic texture), the basic equation for a plane, rotationally symmetric stress state reads [13]:

$$\varepsilon_{\psi}^{hkl} = \left(F_{11}(\psi, hkl) + F_{22}(\psi, hkl)\right)\sigma_{//} \qquad (1)$$

The $F_{ij}(\psi, \varphi, hkl)$ are so-called X-ray stress factors (XSFs; ψ is the inclination angle of the sample surface normal with respect to the diffraction vector). $\sigma_{//}$ can be obtained as the fitting parameter in a least-squares minimisation of the difference of the measured diffraction strains (for a number of different ψ) and the strains calculated using equation (1). The application of equation (1) requires a (separate) establishment of the diffraction elastic constants by adopting a grain-interaction model. This will be discussed next.

THE REUSS, VOIGT AND VOOK-WITT GRAIN-INTERACTION MODELS

For each crystallite, the stress and strain tensors in the specimen frame of reference S, where the S_3-axis is perpendicular to the sample surface, satisfy Hooke's law:

$$\varepsilon_{ij}^{S} = S_{ijkl}^{S}\sigma_{kl}^{S} \qquad (2)$$

The compliance tensor S_{ijkl}^{S} of a crystallite in the S frame depends on the orientation of the crystallite and is calculated from the compliance tensor in the crystal frame of reference by a suitable orthogonal transformation (see, for example, [2,11]). Equation (2) represents a short notation for six equations in twelve unknowns, the stress and strain tensor components (note that the strain and stress tensors are symmetric, i.e. $\varepsilon_{ij} = \varepsilon_{ji}$ and $\sigma_{ij} = \sigma_{ji}$). If six components of the twelve unknowns are known, consequently, the other components can be calculated by solving the system of equations (2). In the type of grain-interaction models considered here, a total of six stress and/or strain tensor components in the S frame are taken equal to the mechanical averages for all crystallites and thus the other six (unknown) components can be calculated from the system of equations (2). Then, the mechanical strain as well as the diffraction strain and the corresponding elastic constants can be calculated by proper averaging over the crystallites (all crystallites for the mechanical constants; selected crystallites for the diffraction elastic constants and the diffraction stress factors) [2].

Among the traditional grain-interaction models, the Reuss, Voigt and Neerfeld-Hill models are relevant to the present work.

In the Voigt model it is assumed that the strain tensor in the S frame is equal for all grains and thus equal to the mechanical strain tensor of the sample whereas in the Reuss model it is assumed that the stress tensor in the S frame is equal for all grains and thus equal to the mechanical stress tensor of the sample. Both models imply unphysical boundary conditions at the grain boundaries.

Hill [6] proved that the mechanical elastic constants according to the Reuss and Voigt models are the theoretical limits (upper and lower bounds) of the elastic constants and suggested on experimental grounds to approximate the mechanical elastic constants by the averages of the Voigt and Reuss values. The same averaging was also proposed earlier on experimental grounds by Neerfeld [5] for the mechanical and diffraction elastic constants. Taking the average of Voigt and Reuss elastic constants is referred to as the Neerfeld-Hill model and, for bulk materials, provides satisfactory values of the mechanical and diffraction elastic constants. A generally accepted physical basis for mixing the models of Reuss and Voigt lacks but a few authors have tried to give an interpretation of such a averaging procedure on a physical basis [12]. The